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Personal exposure to particles in Banská Bystrica, Slovakia[1]

MICHAEL BRAUER,[a] FRANTIŠKA HRUBÁ,[b] EVA MIHALÍKOVÁ,[b] ELEONÓRA FABIÁNOVÁ,[b] PETER MISKOVIC,[b] ALENA PLZIKOVÁ,[b] MARIE LENDACKÁ,[b] JOHN VANDENBERG[c] AND ALISON CULLEN[d,e]

[a]*The University of British Columbia, Vancouver, Canada*
[b]*State Institute of Public Health, Banská Bystrica, Slovak Republic*
[c]*U.S. EPA, Research Triangle Park*
[d]*University of Washington, Seattle*
[e]*National Center for Research in Statistics and the Environment, Seattle*

Epidemiological studies have associated adverse health impacts with ambient concentrations of particulate matter (PM), though these studies have been limited in their characterization of personal exposure to PM. An exposure study of healthy nonsmoking adults and children was conducted in Banska Bystrica, Slovakia, to characterize the range of personal exposures to air pollutants and to determine the influence of occupation, season, residence location, and outdoor and indoor concentrations on personal exposures. Twenty-four-hour personal, at-home indoor, and ambient measurements of PM_{10}, $PM_{2.5}$, sulfate (SO_4^{2-}) and nicotine were obtained for 18 office workers, 16 industrial workers, and 15 high school students in winter and summer. Results showed that outdoor levels of pollutants were modest, with clear seasonal differences: outdoor PM_{10} summer/winter mean = 35/45 $\mu g/m^3$; $PM_{2.5}$ summer/winter mean = 22/32 $\mu g/m^3$. SO_4^{2-} levels were low (4–7 $\mu g/m^3$) and relatively uniform across the different sample types (personal, indoor, outdoor), areas, and occupational groups. This suggests that SO_4^{2-} may be a useful marker for combustion mode particles of ambient origin, although the relationship between personal exposures and ambient SO_4^{2-} levels was more complex than observed in North American settings. During winter especially, the central city area showed higher concentrations than the suburban location for outdoor, personal, and indoor measures of PM_{10}, $PM_{2.5}$, and to a lesser extent for SO_4^{2-}, suggesting the importance of local sources. For $PM_{2.5}$ and PM_{10}, ratios consistent with expectations were found among exposure indices for all three subject groups (personal > indoor > outdoor), and between work type (industrial > students > office workers). The ratio of $PM_{2.5}$ personal to indoor exposures ranged from 1.0 to 3.9 and of personal to outdoor exposures from 1.6 to 4.2. The ratio of PM_{10} personal to indoor exposures ranged from 1.1 to 2.9 and the ratio of personal to outdoor exposures from 2.1 to 4.1. For a combined group of office workers and students, personal $PM_{10}/PM_{2.5}$ levels were predicted by statistically significant multivariate models incorporating indoor (for $PM_{2.5}$) or outdoor (for PM_{10}) PM levels, and nicotine exposure (for PM_{10}). Small but significant fractions of the overall variability, 15% for $PM_{2.5}$ and 17% for PM_{10}, were explained by these models. The results indicate that central site monitors underpredict actual human exposures to $PM_{2.5}$ and PM_{10}. Personal exposure to SO_4^{2-} was found to be predicted by outdoor or indoor SO_4^{2-} levels with 23–71% of the overall variability explained by these predictors. We conclude that personal exposure measurements and additional demographic and daily activity data are crucial for accurate evaluation of exposure to particles in this setting. *Journal of Exposure Analysis and Environmental Epidemiology* (2000) **10,** 478–487.

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Introduction

Although evidence suggests a causal relationship between particulate air pollution and mortality/morbidity, questions remain over the biological basis for such a relationship and the potential misclassification of exposure in epidemiological studies (NRC, 1998, 1999). Current particle air pollution epidemiological studies have generally relied on estimates of exposure in which data from single stationary ambient air pollution monitoring locations are used to represent the exposure of the entire study population. Estimating individual exposure to particles from central outdoor pollution monitors may result in considerable error and can bias the exposure–response relationship. Further, exposure assessment is required to accurately assess population health risks associated with air pollution exposure and to formulate potential exposure reduction policies.

For the most common of the particle epidemiological studies — the time series studies — the use of ambient concentrations as indicators of exposure is based on the

1. Address all correspondence to: Dr. Michael Brauer, School of Occupational and Environmental Hygiene, The University of British Columbia, 2206 East Mall, Vancouver, BC, Canada V6T 1Z3. Tel.: +1-604-822-9585. Fax: +1-604-822-9588.
E-mail: brauer@interchange.ubc.ca
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correlation over time between ambient concentrations and measurements of exposure. Indeed, several studies have measured high correlations over time between ambient concentrations and personal exposures of members of specific subject groups (Janssen et al., 1997a, 1998, 1999; Stieb et al., 1998; Ebelt et al., 2000).

In cross-sectional epidemiological studies of particle air pollution health effects, exposure misclassification may have more important impacts. In these studies, the use of ambient particle measurements as an exposure estimate is justified based on the assumption that ambient monitoring data are adequate surrogates for indoor concentrations, and consequently for personal exposures to particulate matter (PM) of ambient origin. Similarly, these studies assume that a single ambient particle measurement adequately describes spatial variability in ambient concentrations within each of the study areas. Recent studies, however, have provided differing views regarding the importance of spatial variability. For example, a study conducted recently in Eastern Germany suggested that the spatial variability for aerosol sulfate (SO_4^{2-}) and PM_{10}, even in small areas, might be greater than commonly assumed, especially during periods of low wind speed and when the areas are affected by local emissions sources (Cyrys et al., 1998). A study of spatial variability in particle concentrations has shown that $PM_{2.5}$ concentrations near a major road in The Netherlands were 30% greater than at a background location not influenced by local traffic (Janssen et al., 1997b). Black Smoke levels were 2.6 times higher at the roadside locations, indicating the important contribution of diesel exhaust to traffic-related $PM_{2.5}$ emissions. No differences were found for elemental sulfur between the two sites, suggesting a larger influence of regional rather than local pollution sources on sulfur exposures.

Some epidemiological studies have reported differences in respiratory health indicators within cities, supposedly related to differences in traffic density and traffic-related air pollution. Increased respiratory symptoms in children are associated with living near a freeway and with traffic density, especially truck traffic (Van Vliet et al., 1997). Traffic-related differences between neighborhoods have also been studied (Wijst et al., 1993). On a smaller geographical scale, particulate air pollution levels may be high both in and close to streets with a high traffic density, with potential effects on respiratory health of residents living there (Buzorius et al., 1999). This hypothesis has been studied in several locations (Nitta et al., 1993; Duhme et al., 1996; Oosterlee et al., 1996).

In this study, we evaluated several factors that were hypothesized to contribute to personal particle exposures. The measurements were conducted in a risk assessment context, and were made in order to gather new information regarding particle exposures and potential variables affecting exposure within a city in Slovakia. The study was performed in Banska Bystrica, Slovakia, a moderately industrialized city of 85,000 located in the Low Tatras mountain range of central Slovakia. The city center extends along the Hron River with 1000 m mountains located to the immediate north, south, and east of the city center. The population is distributed throughout the city center in homes and apartment buildings of up to eight stories, and in several large blocks of taller, i.e., about 15 stories, apartment buildings. There is a population of approximately 13,000 residents living in the Sasova complex, a suburban residential area of 15-story apartment blocks about 3 km away from the city center and 250 m higher in elevation. Major industries include a large wood processing facility near the city center and a cement plant located 5 km from the city center (Hruba et al., 2000). Local heating sources rely primarily on natural gas. The city center is noted for heavier vehicle traffic relative to the residential area, as well as the presence of several industrial sources, including the wood processing facility. The Sasova area is influenced by local natural gas heating and also by traffic, although less so than the center. In addition, there is a small amount of local heating with coal in one older section of Sasova village.

The cooperative project described in this manuscript was built on data collected as part of the Central European Study on Air Pollution and Respiratory Health (CESAR) study of air pollution and respiratory disease in children (Fletcher et al., 1999). The CESAR study included uniform collection of respiratory health, family health history, household and socio-economic characteristics, and air monitoring data from 25 Central European cities of widely varying air pollution conditions. The objective was to assess, through cross-sectional analyses aggregating responses at the city level air pollution as a risk factor for respiratory disease in children. The CESAR design was intended to compare urban vs. "background" sites and to enable within-country as well as between-country comparisons. It was hypothesized that urban sites would impose higher exposures and supposedly higher respiratory disease health risk than rural sites. We sought to evaluate a related hypothesis regarding exposure differences between residents of urban and background locations; to test this hypothesis, we collected indoor and personal measurements in addition to ambient measurements.

Concurrent objectives of this study were to characterize the range of personal exposures to PM_{10}, $PM_{2.5}$, and SO_4^{2-} for nonsmokers in Banská Bystrica and to evaluate the impact of season and occupational group. We also sought to assess the relationship between personal exposures and indoor and outdoor concentrations. We hypothesized that PM_{10} exposures would be influenced by occupation, indoor exposure, and local variability, suggesting that personal PM_{10} exposures are not accurate measures of personal exposure to PM of ambient origin. Further, we hypothesized that personal $PM_{2.5}$ measurements, while not affected as

much as PM_{10} by spatial variability and by noncombustion particles in the workplace, would still be affected by indoor exposures. SO_4^{2-} was measured to evaluate its potential as a marker for exposure to ambient combustion source particles (Lippmann and Thurston, 1996; Ebelt et al., 2000) while nicotine was measured as a marker of exposure to environmental tobacco smoke (ETS).

Methods

Twenty-four-hour personal exposure measurements were made for PM_{10}, $PM_{2.5}$, SO_4^{2-}, and nicotine for 18 office workers, 15 high school students, and 16 workers from four industrial facilities (cement factory [N=5], wood processing plant [N=6], textile plant [N=4], and bakery [N=1]). Twenty-one of the subjects were male and the remaining 28 female. Subjects were selected from those residing in either the city center or the Sasova residential area. Twenty-four subjects (eight office workers, eight industrial workers, and eight students) lived in the city center and the remaining 25 subjects (nine office workers, eight industrial workers, and eight students) lived in the residential area. The gender distribution of subjects was matched within each occupational category when split by residential location. Earlier studies of ambient air pollution in both areas had suggested that ambient levels of PM were significantly lower in the residential area than the city center. The annual average of PM_{10} was 47 as opposed to 40 $\mu g/m^3$, while $PM_{2.5}$ levels were 34 as opposed to 29 $\mu g/m^3$ for the city center and the Sasova residential area, respectively (Mihalková et al., 1998). All subjects living in the Sasova area resided in large apartment buildings, while subjects from the city center area lived in similar apartment buildings (45%), semi-detached homes (40%), or detached homes (15%).

Samples were collected during both summer (June 17–September 18, 1997) and winter (November 17, 1997–March 10, 1998) periods; each subject had one 24-h measurement made during each season. In addition to personal samples, indoor measurements were made in the homes of all participants and outdoor samples were collected in each of the two study areas on each day of measurement. Two subjects (generally from the same residential location) were monitored on each day during each of the sampling periods. Indoor sampling was conducted in the room where people reported they spent the majority of their time when not sleeping. Outdoor sampling was conducted at sites previously selected for a European community collaborative research project (Fletcher et al., 1999). These sites were selected to be "background" sites (not influenced by local sources) that were representative of the surrounding community. Outdoor samples were collected at a height of approximately 2 m above ground level. Both indoor and outdoor samples were collected at a distance of at least 1 m from all vertical surfaces (walls). The indoor samples were collected at a height of approximately 1.5 m. In total, 95 measurements (2 measurements per person×49 persons; three subjects did not participate in the winter sampling period) were made. Of these 95 possible measurements, 98–100% (for the various samples: indoor, outdoor, personal, PM_{10}, $PM_{2.5}$, SO_4^{2-}) was actually collected. Of these samples, valid analytical results were obtained for 87, 94, and 82 samples for indoor, outdoor, and personal PM_{10}, respectively; 87, 92, and 81 for indoor, outdoor and personal $PM_{2.5}$, respectively; 87, 94, and 82 for indoor, outdoor and personal SO_4^{2-}, respectively. These represent valid sample collection efficiencies between 85% and 100%. Samples were excluded if they were statistical outliers and there was evidence of obvious errors (pump failures, battery failures, contaminated filters, etc.) based on laboratory and field notes.

Quality control was assessed by use of quality control charts to monitor systematic changes in mass or SO_4^{2-} concentrations of a series of control filters. Approximately 10% of samples was composed of field blanks. Spiked filters and field blanks were also used to estimate the measurement repeatability and reproducibility. For PM mass, the mean repeatability standard deviation (repeated measurements of the same filters by the same analyst on the same day) was 7.9 μg, corresponding to an estimated concentration repeatability of 2.7 $\mu g/m^3$, assuming a 24-h sample collected at 2 l/min. The mean PM mass reproducibility standard deviation (repeated measurements of the same filters by different analysts on different days) was 15.0 μg, corresponding to an estimated concentration reproducibility of 5.37 $\mu g/m^3$, assuming a 24-h sample collected at 2 l/min. Repeatability and reproducibility could not be assessed for SO_4^{2-} since the entire extract volume had to be used for capillary zonal electrophoresis analysis.

Outdoor particle samples ($PM_{10/2.5}$) were collected with Harvard Impactors (Air Diagnostics and Engineering Inc., Naples, Maine, USA, operated at a flow rate of 10 l/min). Indoor and personal samples were collected at flow rates of 2 l/min with PM_{10} and $PM_{2.5}$ Personal Environmental Monitors (MSP Corp.) connected to personal sampling pumps (Universal PCXR4, SKC Inc.) which used additional battery packs (P21661, SKC Inc.) to operate uninterrupted for 24 h. The flow from the sampling pump was split into two 2 l/min sampling flows by an adapter plug and was checked at both arms simultaneously. Sampling flow rates were measured with rotameters that were calibrated with an NIST traceable "frictionless" piston flow meter (Drycal DC-1, Bios Corp.).

Samples were collected on Teflon membrane filters (Teflon, Gelman Sciences) and mass concentrations were determined with a microbalance (Mettler-Toledo AE 240) with a sensitivity of 20 μg. The detection limits of the

gravimetric analysis were 25 and 5 $\mu g/m^3$ for personal/indoor and outdoor samples, respectively. The LOD is based on 3 SD of the mass gain of laboratory blanks and a sample flow of 2.9 m^3 for personal/indoor samples and 14 m^3 for outdoor samples. The high LOD for personal/indoor samples is due to the low sample volume and low sensitivity of the balance. No outdoor samples were below the LOD. 99, 97, 92 and 80% of personal PM_{10}, personal $PM_{2.5}$, indoor PM_{10} and indoor $PM_{2.5}$ samples, respectively, were above the LOD. All valid samples, including those below the LOD, were used (as measured values) in the statistical analyses.

SO_4^{2-} concentrations from the PM_{10} and $PM_{2.5}$ filters were measured by capillary zonal electrophoresis following ultrasonic extraction in distilled, deionized water. The capillary zonal electrophoresis method uses a hydrodynamically closed separation system with conductivity detection. An Isotachophoretic analyzer (Villa-Labeco, Slovakia) was used in the single-column configuration of the separation unit. The length of the capillary tube was 250 mm. The composition of the carrier electrolyte was 7 mmol/l succinate acid +0.5 mmol/l bis-tri-propan, 5% (w/v) (polyvinylpyrrolidone, 0.1% w/v) methyl hydroxy ethylcellulose at pH=3.55 (Kaniansky et al., 1996). The LOD of the SO_4^{2-} analysis was 1.8 $\mu g/m^3$ based on a sample flow of 2.9 m^3 and estimation of the method detection limit from the peak-to-peak noise ratio as described by Foley and Dorsey (1984). All samples were above the detection limit.

For indoor and personal samples, the Teflon filters were backed by a polypropylene separator (see following paragraph) (Gelman Sciences) and a sodium bisulfate impregnated glass fiber filter (Gelman Sciences) for the collection of nicotine. Glass fiber filters were impregnated by coating with an aqueous solution of 4% sodium bisulfate. For coating, each filter was placed on the surface of the solution for a few seconds, until it had absorbed the solution, and then the filter was placed on a clean glass plate to dry. In this way, each filter was coated with 7–10 mg sodium bisulfate. Nicotine was determined by gas chromatography/mass spectrometry using ketamine as an internal standard (Hammond et al., 1987). The nicotine detection limit of 0.2 $\mu g/m^3$ was based on 3 SD of the nicotine measured on laboratory blanks and a sample flow of 2.9 m^3 for personal/indoor samples.

During the first measurement period (summer 1997), we noticed that the sodium bisulfate-coated backup filters (used to collect nicotine) resulted in SO_4^{2-} contamination of the Teflon filters. This invalidated the personal and indoor SO_4^{2-} results from the summer measurement period. All subsequent measurements were made with the use of a polypropylene separator between the Teflon and coated filters. We also evaluated whether the coated filter was affecting the gravimetric measurements by performing a series of experiments using coated filters with and without back-up filters. On average, the coated filter without separator led to a 67 μg (SD=24 μg) increase in Teflon filter mass. This value was then used to correct the mass measurements for all Teflon filters from the summer 1997 period. A positive mass gain of 17 μg was evident from blank Teflon filters for the winter 1997 sampling period, and all mass samples from this period were corrected by subtracting this amount. The average mass gains on all sample filters were 250, 530, and 425 μg for indoor, outdoor and personal PM_{10}, respectively, and 182, 355, and 285 for indoor, outdoor and personal $PM_{2.5}$, respectively. Note that the outdoor samples collected greater mass due to the higher flow rate (10 l/min) relative to the indoor and personal samples (2 l/min).

In addition, time–activity data and housing characteristics information were collected by 24-h recall interviews with study subjects. All interviews were conducted by trained field staff using a standardized questionnaire format. Participants were free to report time–activity flexibly and without the imposition of pre-set time period resolution. We evaluated the following variables as predictors of personal PM and SO_4^{2-} exposure: ambient and home indoor concentrations of PM/SO_4^{2-}, personal nicotine exposure, type of work, time spent at home/at work/in transit, location of residence, time spent cooking, and self-reported exposure to ETS.

Results and discussion

Summary statistics of time spent in different microenvironments (Table 1) indicated that during sample collection periods, participants spent nearly all of their time either at home (mean=17 h/day; range 10–24 h/day) or at work/school (mean=6 h/day; range 0–13 h/day). The mean time spent in transport was 13 min/day, and participants spent an average of 36 min/day cooking. Only a small number of participants reported having spent any time outdoors during sampling. This suggests that there was substantial activity modification during sampling and that subjects may have avoided spending time outdoors or in public places (Boudet et al., 1997). Another explanation is underreporting by participants of time spent outdoors and in

Table 1. Average time spent in each microenvironment.

	Students	Office workers	Industrial workers	All groups
Outside	13 (49)	1.2 (8)	36 (92)	14 (55)
Home	1191 (226)	1005 (141)	923 (147)	1034 (197)
Work	209 (194)	385 (138)	457 (86)	356 (172)
Cooking	38 (40)	36 (38)	34 (57)	36 (44)
Travel time	9 (10)	14 (8)	16 (9)	13 (9)

Data from both summer and winter measurement periods in minutes per day (standard deviation).

transit. Seasonal differences in time–activity patterns also were observed. Students reported large increases in time spent at home during the summer and corresponding decreases in time spent at work. All groups reported increased outdoor time during the summer. Forty-five out of 46 participants reported the presence of a gas stove/oven at their home while only seven out of 46 (two students, five office workers) reported the presence of a gas heater or exposure to ETS. Nicotine concentrations were very low; 31% of personal samples and 52% of indoor samples were below the detection limit of 0.2 $\mu g/m^3$. However, personal exposure to nicotine was measured even for individuals who reported no exposure to ETS. This result indicates the importance of collecting objective exposure measurements to assess ETS exposure.

Indoor:Outdoor Relationships

Indoor:outdoor ratios were greater than 1 for PM_{10}, $PM_{2.5}$ and less than 1 for SO_4^{2-}. No statistically significant seasonal differences in indoor:outdoor ratios were observed, but both PM_{10} and $PM_{2.5}$ indicated the same tendency for higher indoor:outdoor ratios in summer than in winter. Geometric mean indoor:outdoor ratios during summer were 1.9 and 1.8 for PM_{10} and $PM_{2.5}$, respectively, and 1.5 and 1.6 during winter. The winter geometric mean indoor:outdoor SO_4^{2-} ratio was 0.8. No differences were observed between the downtown and residential areas. Consistent with other studies, these findings suggest indoor sources of PM_{10} and $PM_{2.5}$. While indoor sources of PM_{10} and $PM_{2.5}$ are known (resuspended dust, ETS, cooking emissions), major indoor SO_4^{2-} sources have not been recognized to date.

Personal:Indoor, Personal:Outdoor Ratios

As shown in previous studies (Lioy et al., 1990; Özkaynak et al., 1996; Janssen et al., 1997a, 1998, 1999; Ebelt et al., 2000), personal:indoor and personal:outdoor concentration ratios were greater than 1 for all demographic groups for PM_{10} and $PM_{2.5}$ (Tables 2 and 3). For SO_4^{2-}, personal:indoor and personal:outdoor ratios were also greater than 1 for the students and industrial workers, with ratios slightly below 1 for the office workers. While this was expected for PM_{10} and $PM_{2.5}$, the finding for SO_4^{2-} was unexpected as SO_4^{2-} is believed to have no major indoor sources or "personal cloud". For PM_{10} and $PM_{2.5}$, but not for SO_4^{2-}, personal:outdoor ratios were greater than corresponding personal:(home) indoor ratios as expected, given that nearly all participants spent most of the sampling period indoors. This also suggests that the elevated personal exposures were due, in part, to indoor sources. Summer personal:indoor and personal:outdoor ratios were higher than those for winter for PM_{10} and $PM_{2.5}$ (confirmed by factorial ANOVA). This may reflect the impact of at-home activities which were more common in the summer than during winter.

For all three subject groups, personal exposures were greater than indoor or outdoor levels. The group of factory workers had significantly higher ratios and exposures than the other groups (Figures 1a–c) for PM_{10} and $PM_{2.5}$, but not SO_4^{2-}. This is consistent with occupational exposures to particles and partially supports our hypothesis that SO_4^{2-} could be used as a particle marker which is not affected by occupational exposure.

SO_4^{2-} is thought to have no major indoor sources and to reflect regional particulate air pollution. As geometric mean personal:indoor SO_4^{2-} ratios were also above 1 and geometric mean indoor:outdoor SO_4^{2-} ratios were below 1, the elevated personal SO_4^{2-} exposures cannot be explained by indoor residential SO_4^{2-} exposure. Further, as indoor samples were collected from homes and since a major portion of the day was spent outside the home, these elevated ratios for SO_4^{2-} may be due to exposures experienced while away from home. One possible explanation consistent with our findings is that SO_4^{2-} exposures were influenced by local sources which were not well characterized by the ambient monitoring locations. Since the SO_4^{2-} levels in all measurements were quite low, averaging about 6 $\mu g/m^3$, it is also possible that even minor contamination of the Telfon filters by the sodium bisulfate glass fiber filter material could have had an inordinate effect on the calculated ratios. However, as both personal and indoor samples would have been subject to any such effect, this is unlikely to explain the elevated personal:indoor

Table 2. Geometric mean personal:indoor ratios by season and subject group.

	PM_{10}		$PM_{2.5}$		SO_4^{2-}	
	Summer	Winter	Summer	Winter	Summer	Winter
Students	1.1	2.1	1.1	1.2	–	1.5
Office workers	1.8	1.1	2.6	1.0	–	0.98
Industrial workers	2.9	2.6	3.9	1.7	–	1.8

Summer SO_4^{2-} indoor and personal samples were contaminated and are not reported. Winter SO_4^{2-} samples were not subject to contamination due to use of separator between Teflon and coated filter (see description in Methods).

Table 3. Geometric mean personal:outdoor ratios by season and subject group.

	PM_{10}		$PM_{2.5}$		SO_4^{2-}	
	Summer	Winter	Summer	Winter	Summer	Winter
Students	2.7	2.7	3.3	1.6	–	1.4
Office workers	3.1	2.1	3.7	1.9	–	.82
Industrial workers	4.1	3.3	4.2	3.4	–	1.2

Summer SO_4^{2-} indoor and personal samples were contaminated and are not reported. Winter SO_4^{2-} samples were not subject to contamination due to use of separator between Teflon and coated filter (see description in Methods).

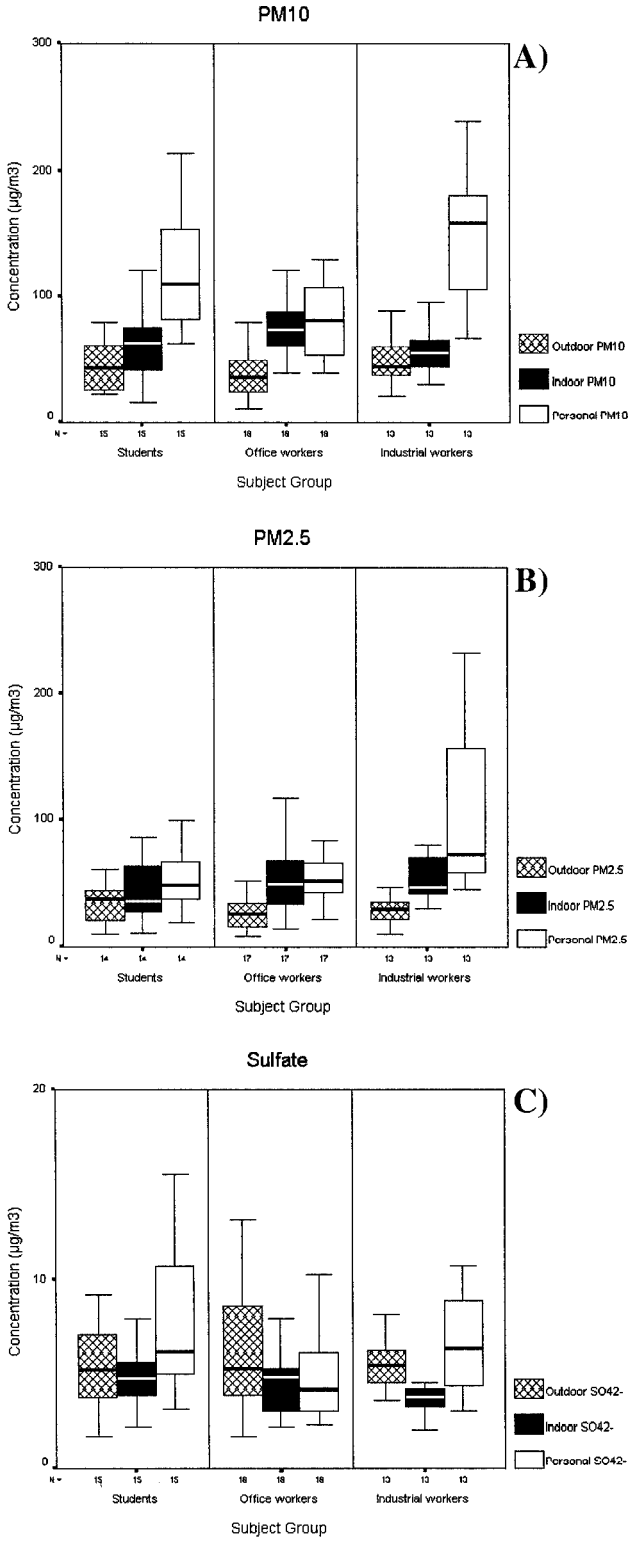

Figure 1. Distributions of outdoor, indoor, and personal (A) PM_{10}, (B) $PM_{2.5}$ and (C) SO_4^{2-} concentrations for each subject group. Samples from winter period only are presented.

ratios. Use of a polypropylene separator between the coated and Teflon filters, however, resulted in no measurable SO_4^{2-} contamination based on analysis of filter blanks.

Seasonal Differences

As expected, based on local meteorology and increased use of heating, outdoor PM_{10} and $PM_{2.5}$ measurements were significantly higher during winter than in summer. In contrast, outdoor SO_4^{2-} measurements indicated similar (for the center) and even slightly higher (for the residential area) summer concentrations, likely due to increased regional SO_4^{2-} levels from summer photochemistry. For PM_{10} and $PM_{2.5}$, both indoor and personal exposure measurements were similar or slightly higher in summer than in winter (Table 4) — a pattern that did not correspond to outdoor levels. One possible explanation is that time–activity patterns were quite different between the winter and summer measurement periods. During the summer measurement period, the majority of measurements were made during the July–August school vacation period, when more people were at home, undertaking more potential particle-generating activities during the day (cleaning, gardening, etc.). During the winter sampling period, most homes were empty during working hours. Due to these clear seasonal differences, all analyses are presented during both seasons.

Residential Location

For PM_{10}, $PM_{2.5}$, and SO_4^{2-}, indoor and outdoor levels from the downtown area were higher than in the residential area (Table 5). These differences, however, were not always reflected in the personal exposure measurements, particularly during the summer period. The finding of spatial variability in PM_{10}, $PM_{2.5}$, and, to a lesser degree, SO_4^{2-} outdoor levels suggests that the observed spatial differences in indoor PM_{10} and $PM_{2.5}$ levels are due to different concentrations of ambient particles (and their penetration indoors) and not solely due to differential effects of indoor sources between the two areas. There did not appear to be a strong influence of outdoor concentration spatial variability on personal exposure. When data from both seasons were combined, the only significant differences observed between areas were for outdoor measurements. Together, these results indicate higher outdoor particle levels in the downtown area of the city. This is also reflected in higher indoor levels, particularly in the summer, but not in higher levels of personal exposure. During the winter, however, the spatial variability does appear to result in higher personal exposures for those residing downtown. The fact that these patterns are also observed for SO_4^{2-} measurements suggests that they are due to differences in ambient concentrations and not to the presence of indoor or occupational sources. One limitation of our study design was that observed differences between locations may be biased due to meteorological factors

Table 4. Summary statistics of measured concentrations ($\mu g/m^3$) of PM_{10}, $PM_{2.5}$, and SO_4^{2-} by season and sample type.

	Summer		Winter	
	Arithmetic mean (SD)	Geometric mean (GSD)	Arithmetic mean (SD)	Geometric mean (GSD)
PM_{10} ($\mu g/m^3$)				
Outdoor (N=48/46)	35 (11)	33 (1.4)	45 (20)	40 (1.6)*
Indoor (N=41/46)	79 (53)	63(2.0)	66 (27)	60 (1.6)
Personal (N=36/46)	122 (65)	107 (1.7)	120 (74)	105 (1.7)
$PM_{2.5}$ ($\mu g/m^3$)				
Outdoor (N=48/44)	22 (7)	21 (1.4)	32 (16)	27 (1.7)*
Indoor (N=41/46)	55 (52)	37 (3.0)	53 (24)	47 (1.6)
Personal (N=35/46)	88 (44)	77 (1.7)	69 (43)	60 (1.7)*
SO_4^{2-} ($\mu g/m^3$)				
Outdoor (N=48/46)	6.7 (3.1)	6.0 (1.7)	5.7 (2.6)	5.1 (1.6)
Indoor (N=0/46)			4.6 (2.0)	4.2 (1.5)
Personal (N=0/46)			6.5 (3.9)	5.6 (1.7)

*Indicates significant difference in geometric means at $p<0.05$ for seasonal comparison. Summer SO_4^{2-} indoor and personal samples were contaminated and are not reported. Winter SO_4^{2-} samples were not subject to contamination due to use of separator between Teflon and coated filter (see description in Methods). N indicates number of valid samples collected during summer/winter periods.

which were not controlled for in either the data analysis or the study design.

The observation of spatial variability in ambient SO_4^{2-} levels was somewhat surprising, although consistent with at least two other studies from Europe. The spatial variability of aerosol SO_4^{2-} was investigated at five sites in Eastern England (Kitto and Harrison, 1992) and more recently for The Netherlands (Hoek et al., 1996). The authors found that the SO_4^{2-} concentration at the urban site was about 10% higher than the concentration at a semi-rural site, located about 60 km away from the urban site. The authors explain the small spatial variability with the long-range transport characteristic for these components due to a slow conversion of SO_2 to SO_4^{2-} and deposition rate. Our measurements of ambient SO_4^{2-} of 5–7 $\mu g/m^3$ agree well with recent estimates for regional concentrations throughout Central Europe (WHO, 1999).

Type of Work

Differences were observed between the three groups of subjects in terms of personal exposures to PM_{10} and $PM_{2.5}$ (Figures 1a–c), with the industrial workers experiencing higher exposures than the office workers or students. These differences were present during both seasons, although figures are only presented for the winter. These workers also had higher personal:indoor and personal:outdoor ratios than the other groups, suggesting that these elevated exposures were, in fact, due to occupational exposures. This interpretation is strengthened by our finding of no significant differences in indoor–home or outdoor levels for the three groups of subjects. No significant differences in exposures to SO_4^{2-} were observed between the three groups, indicating that SO_4^{2-} may be a better marker of exposure to ambient particles than personal measurements of PM_{10} or $PM_{2.5}$, which appear to be affected by occupational exposures. No statistically significant differences were observed between the students and office workers.

Determinants of Exposure

Regression analysis was used to identify determinants of personal exposure to $PM_{2.5}$, PM_{10}, and SO_4^{2-}. Models predicting personal exposure were tested for the group of industrial workers, a combination of office workers and students, and a combination of all three participant groups. Models hypothesizing the relationship between personal exposure and the predictors: indoor and outdoor concentrations, type of work, time spent at home/at work/in transit, location of residence, time spent cooking, self-reported exposure to ETS, and personal nicotine exposure were developed for PM_{10}, $PM_{2.5}$ and SO_4^{2-}. Models were constructed by including all variables together in every model and using a stepwise regression procedure to select variables that were significant predictors. Correlations among model parameters were also evaluated. The only variables that were highly correlated with each other ($r>0.2$) were time spent at home/at work/in transit. This was expected, as these are mutually exclusive. As there was no *a priori* reason for excluding one or the other of these variables, they were all allowed to enter the regression and to allow the stepwise procedure perform the variable selection. Since models that included all subjects simultaneously indicated that the type of work was a significant predictor (with industrial workers being significantly

Table 5. Summary statistics of measured concentrations ($\mu g/m^3$) of PM_{10}, $PM_{2.5}$, and SO_4^{2-} by location and sample type.

	Suburban		Downtown	
	Arithmetic mean (SD)	Geometric mean (GSD)	Arithmetic mean (SD)	Geometric mean (GSD)
Summer				
PM_{10} ($\mu g/m^3$)				
Outdoor (N=24/24)	32 (11)	30 (1.4)	38 (10)	37 (1.3)*
Indoor (N=22/19)	69 (38)	58 (1.9)	90 (66)	69 (2.2)
Personal (N=21/15)	131 (71)	117 (1.6)	109 (56)	94 (1.8)
$PM_{2.5}$ ($\mu g/m^3$)				
Outdoor (N=24/24)	20 (7)	18 (1.5)	25 (7)	24 (1.3)*
Indoor (N=22/19)	46 (31)	33 (2.9)	67 (67)	41 (3.2)
Personal (N=21/14)	89 (44)	80 (1.6)	86 (44)	73 (2.0)
SO_4^{2-} ($\mu g/m^3$)				
Outdoor (N=24/24)	5.7 (2.5)	5.1 (1.7)	7.8 (3.4)	7.0 (1.6)*
Indoor				
Personal				
Winter				
PM_{10} ($\mu g/m^3$)				
Outdoor (N=21/25)	38 (16)	36 (1.4)	50 (23)	44 (1.8)
Indoor (N=21/25)	57 (18)	54 (1.4)	74 (31)	67 (1.7)
Personal (N=21/25)	110 (56)	99 (1.6)	129 (86)	110 (1.7)
$PM_{2.5}$ ($\mu g/m^3$)				
Outdoor (N=21/23)	27 (14)	24 (1.7)	36 (17)	32 (1.7)
Indoor (N=21/25)	41 (15)	39 (1.5)	62 (26)	56 (1.7)*
Personal (N=21/25)	58 (37)	50 (1.7)	78 (46)	69 (1.6)*
SO_4^{2-} ($\mu g/m^3$)				
Outdoor (N=21/25)	5.2 (2.3)	4.7 (1.6)	6.1 (2.8)	5.6 (1.6)
Indoor (N=21/25)	4.3 (1.4)	4.1 (1.4)	4.8 (2.3)	4.3 (1.6)
Personal (N=21/25)	6.0 (3.1)	5.4 (1.6)	6.9 (4.5)	5.8 (1.8)

*Indicates significant difference in geometric means at $p<0.05$ for downtown residential comparison. Summer SO_4^{2-} indoor and personal samples were contaminated and are not reported. Winter SO_4^{2-} samples were not subject to contamination due to use of separator between Teflon and coated filter (see description in Methods). N indicates number of valid samples collected from suburban/downtown locations.

different than the other two groups), we constructed separate models (now excluding type of work) for the industrial workers and for a combined group of students and office workers.

Of the overall variability in personal PM_{10} exposures for the students and office workers, 17% was explained by regression models including outdoor PM_{10} and nicotine as significant predictors; all other variables were not significant. For personal $PM_{2.5}$, indoor $PM_{2.5}$ was the only significant predictor in the model which explained 15% of the overall variability. Personal exposure to SO_4^{2-} was predicted (adjusted R^2=0.23) by a model with outdoor SO_4^{2-} as the only significant variable. The proportion of variance explained by this model is markedly lower than other assessments of personal SO_4^{2-} exposure (Brauer et al., 1989; Suh et al., 1992; Ebelt et al., 2000). For industrial workers, a model predicting personal PM_{10} exposures included travel duration as the only significant variable and explained 46% of the overall variability. For $PM_{2.5}$ exposures of the industrial workers, no models were predictive. Personal exposure to SO_4^{2-} was predicted well (adjusted R^2=0.71) by a model with indoor SO_4^{2-} as the only significant variable.

Discussion

In summary, the ambient levels of pollutants measured in Banska Bystrica, Slovakia, were relatively modest: PM_{10} outdoor ~30–50 $\mu g/m^3$, $PM_{2.5}$ outdoor ~20–35 $\mu g/m^3$,

SO_4^{2-} outdoor ~4–7 $\mu g/m^3$. Our findings for ratios and relationships between indoor, outdoor, and personal measures were generally consistent with expectations. For PM_{10} and $PM_{2.5}$, personal exposures were greater than corresponding indoor or outdoor measurements. While differences were observed for type of work, the general trend was consistent, suggesting that the observed "personal cloud" is a general phenomenon that may relate to a broad range of normal activities (Wallace, 1996). As expected, exposures differed by demographic group. Factory workers with occupational exposures to particles had measurably higher exposures to PM_{10} and $PM_{2.5}$ than either office workers or students. In contrast, SO_4^{2-} levels did not appear to be affected as much by the type of work of the subject and therefore, may be a better indicator of exposure to ambient particles. While indoor SO_4^{2-} levels were slightly lower than outdoor levels, in some cases, the personal exposures to SO_4^{2-} were higher than either indoor or outdoor levels. In contrast to other settings where personal SO_4^{2-} exposures have been measured, ambient SO_4^{2-} concentrations explained a much smaller proportion of the variability in personal exposures. This finding suggests the presence of unrecognized sources of SO_4^{2-} exposure that are not adequately characterized by ambient or indoor monitoring. The observation of local-scale spatial variability in outdoor SO_4^{2-} concentrations also suggests that local sources such as coal burning or diesel emissions may be important contributors to exposure.

As expected, outdoor PM_{10} and $PM_{2.5}$ measurements were significantly higher during winter than in summer. In contrast, outdoor SO_4^{2-} levels were higher in summer than in winter. Subjects residing in the downtown area experienced higher exposures to PM_{10}, $PM_{2.5}$, and SO_4^{2-} than did subjects residing in the residential area. The explanation for this result that is most consistent with our observations is true differences in ambient exposure, although it is not possible to rule out other potential explanations such as the presence of differential indoor penetration of particles or the effect of unknown indoor particle sources. It should be noted that the subjects residing in the residential area all live in high-rise apartment buildings that are nearly identical. Downtown area housing is much more heterogeneous.

Statistically significant relationships were found between personal exposures to PM or SO_4^{2-} and predictors including indoor and outdoor levels, nicotine levels, time–activity patterns, and area of residence. A small fraction of the overall variability, 26% for $PM_{2.5}$ and 26% for PM_{10}, was explained over the combined group of study participants. Personal exposure to SO_4^{2-} was predicted by regression models for individual worker groups and for a combined group, with 23–71% of the overall variability explained.

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